

Evolution toward Quantum Critical End Point in UGe_2

Hisashi KOTEGAWA^{1,2*}, Valentin TAUFOR², Dai AOKI², Georg KNEBEL², Jacques FLOUQUET²

¹*Department of Physics, Kobe University, Kobe 657-8501, Japan*

²*INAC/SPSMS, CEA-Grenoble, 17 rue des Martyrs, 38054 Grenoble, France*

We report on Hall resistivity and electrical resistivity measurements under pressure and magnetic field in UGe_2 with ferromagnetic (FM) tricriticality. The Hall resistivity sensitively detects the first order metamagnetic transition from a paramagnetic (PM) phase to a FM phase in a large pressure range almost up to the quantum critical end point (QCEP). The drastic change in the Fermi surface at the PM-FM transition is detected up to the vicinity of the QCEP, while a strong modification in the field variation of the inelastic scattering between electrons is observed toward the QCEP. The comparison with the theoretical predictions is made.

KEYWORDS: quantum critical end point, UGe_2 , metamagnetism, Hall effect

The Curie temperature T_{Curie} of a paramagnetic (PM) - ferromagnetic (FM) phase transition can be driven to 0 K by applying a critical pressure P_C . In itinerant FM systems, the PM-FM transition goes from second order to first order at the tricritical point (TCP) which is at a pressure below P_C .¹⁻⁶ One consequence of the change of order at the TCP is that, above P_C and at low temperature, the first order PM-FM transition is induced by the magnetic field along the easy magnetization axis. At higher temperature, this first order transition changes to a crossover through a critical end point at T_{CEP} . The line of T_{CEP} under pressure and magnetic field defines FM wings.^{4,5} Such behavior would not be observed for a second order phase transition since the applied magnetic field in itself breaks the time reversal symmetry. On increasing pressure, the field induced first order PM-FM transition terminates at a quantum critical end point (QCEP) at 0 K which is characterized by its pressure P_{QCEP} and its field H_{QCEP} . Thus T_{CEP} starts at T_{TCP} at $H = 0$ and ends up at H_{QCEP} at $T = 0$ K. The QCEP is clearly distinguished, by the lack of spontaneous symmetry breaking, from a conventional quantum critical point which is a second order phase transition at 0 K. The QCEP is a fascinating target for theories⁵⁻¹⁰) but experimentally represents a challenge due to the lack of systems which can be studied at accessible field and pressures. Here as the FM wing is drawn almost up to QCEP, comparison can be made with theoretical proposals.

UGe_2 is a unique example as the conventional itinerant FM limit ($P_C \gg 0$) has been precisely determined and it is possible to produce high quality single crystals. Furthermore, UGe_2 has been already extensively studied as it is the first discovered ferromagnetic superconductor.¹¹) At ambient pressure, the Curie temperature ($T_{\text{Curie}} \sim 52$ K) is associated with the large sublattice magnetization $M_0 \sim 1.5 \mu_B$. With increasing pressure, the magnetic ground state switches from a highly polarized phase (FM2, $M_0 \sim 1.5 \mu_B$) to a weakly polarized phase (FM1, $M_0 \sim 0.9 \mu_B$) at a pressure $P_x \sim 1.2$ GPa

through a first order transition. Further increasing the pressure, the FM1 phase collapses at a critical pressure $P_C \sim 1.5$ GPa with an abrupt drop of the sublattice magnetization $\Delta M_0 = 0.9 \mu_B$.¹²) De facto, it is this large drop of ΔM_0 which allows one to observe the pressure and magnetic field evolution of the transitions over a large pressure-field window up to QCEP. Recently the TCP at zero field has been observed by thermal expansion measurements³) and resistivity measurements which indicate $P_{\text{TCP}} = 1.42$ GPa and $T_{\text{TCP}} = 24$ K.⁴)

Here we report the pressure and field evolution of PM-FM1 transition by Hall and resistivity measurements up to 3.41 GPa in UGe_2 . In our previous experiments,⁴) we reported the observation of the TCP and the (P, T, H) phase diagram with the first order plane. However, the pressure was much lower than P_{QCEP} . Other previous measurements were also limited to 2 GPa.¹²⁻¹⁵) In the present experiments we were able to approach close to the QCEP and demonstrate the clear separation between the first order transition and crossover regime by Hall measurements which have the benefit of being sensitive to changes in Fermi surface (FS).

A single crystal of $0.8 \times 0.3 \times 0.15 \text{ mm}^3$ was used and was prepared as previously described.⁴) The Hall resistivity ρ_{xy} and electrical resistivity ρ_{xx} were measured using a four-probe AC method. The electrical current was applied along the c -axis in the orthorhombic structure. ρ_{xy} was extracted from the difference between positive and negative field, namely $(\rho_{H+} - \rho_{H-})/2$, while the ρ_{xx} was obtained by the average, namely $(\rho_{H+} + \rho_{H-})/2$. Pressure was applied up to 3.41 GPa using an indenter cell with Daphne7474 as a pressure-transmitting medium.^{16,17}) The pressure was determined by the superconducting transition temperature of lead. For low pressures, up to 2.72 GPa, the measurements were performed using Quantum Design Physical Properties Measurement System (PPMS) with a maximum field of 9 T and a base temperature of 1.8 K, while for higher pressures a conventional dilution refrigerator was used with a maximum field of 16 T and a base temperature of 0.22 K.

Figure 1 shows the field dependence of ρ_{xx} and ρ_{xy} for several temperatures at 2.25 GPa, where the ground

*E-mail address: kotegawa@crystal.kobe-u.ac.jp

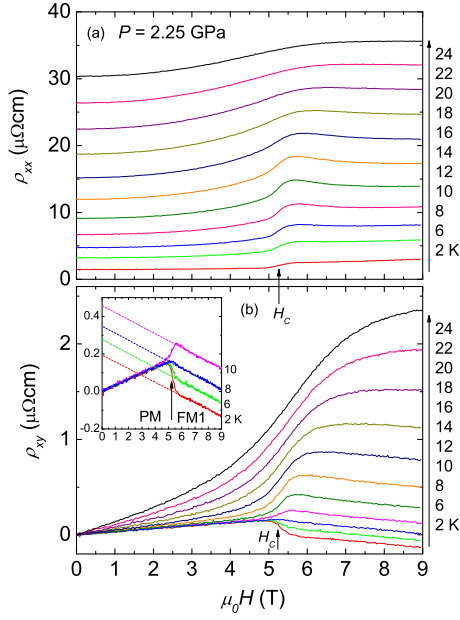


Fig. 1. (color online) Field dependence of (a) ρ_{xx} and (b) ρ_{xy} for $H \parallel a$ -axis at 2.25 GPa. The transitions at H_C are of first order at low temperatures, and change to a crossover at high temperatures. The inset in lower panel shows ρ_{xy} at low temperatures. The finite intercept extrapolated from the FM1 phase above H_C confirms that FM1 phase has a spontaneous magnetization.

state of UGe_2 is PM at zero field. When the magnetic field is applied along the a axis (easy magnetization axis) at low temperature, the system shows the metamagnetic transition from the PM phase to the FM1 phase at H_C accompanied by a step-like jump in ρ_{xx} and ρ_{xy} . At higher temperatures, the PM–FM1 transition changes to a crossover.⁴⁾ In both, ρ_{xx} and ρ_{xy} , the sharp anomaly appears at $H_C \sim 5.2$ T at low temperatures, but it is smeared out at high temperatures. The inset of Fig. 1(b) shows ρ_{xy} between 2 and 10 K. The field dependence of ρ_{xy} is almost linear for both the PM and the FM1 phase. In general, ρ_{xy} is expressed as follows,

$$\rho_{xy} = R_0\mu_0H + R_s\mu_0M, \quad (1)$$

where M is the magnetization. The first term is attributed to the ordinary Hall effect related to the carrier density and the carrier mobility. The second term is the anomalous Hall effect originating from skew scattering and side-jump scattering, and it has been reported that the relation of $R_s \propto \rho_{xx}^2$ is dominant in the FM state of UGe_2 .¹⁸⁾ At low temperatures, no strong field dependence of ρ_{xx} is observed in either the PM or the FM1 phase hence the linear field dependence of ρ_{xy} observed in these phases which indicates that field dependence of the carrier density is small. The sign change in the slope between FM1 and PM implies a change in the FS with opposite signs for the dominant carrier. At ambient pressure, band structure calculations have shown that there is a topological change at the FS of UGe_2 , which is a compensated metal, between 2 hole bands and 2 electron bands in the FM phase and 2 hole bands and 1 electron band in the PM phase.¹⁹⁾ The intercept of ρ_{xy} extrapolated to $H = 0$ corresponds to $R_s\mu_0M(H \rightarrow 0)$.

This term is zero in the PM state while it has a nonzero value in the extrapolation from the FM1 phase, indicative of a spontaneous magnetization. The quantitative analysis is difficult due to the lack of absolute magnetization data under pressure, but the Hall effect is found to be a good tool to detect the PM–FM transition through the changes in both the FS and in M .

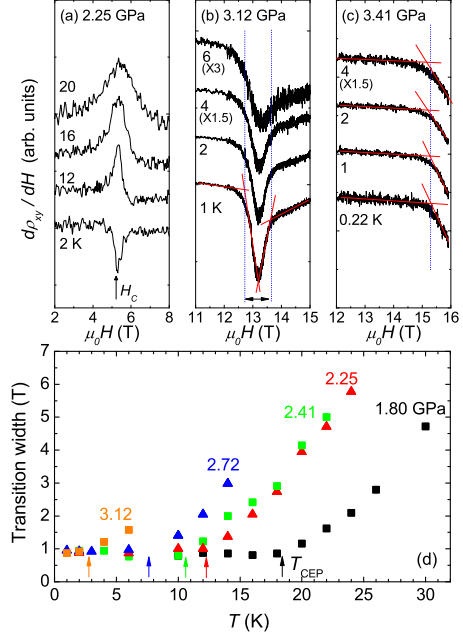


Fig. 2. (color online) Field dependence of $d\rho_{xy}/dH$ at (a) 2.25, (b) 3.12, and (c) 3.41 GPa. H_C was deduced from the peak in $d\rho_{xy}/dH$. The transition width was estimated from the cross point of the linear extrapolations. (d): Temperature dependence of the transition width in the field sweep. The width is kept as sharp as ~ 1 T in the first-order transition, but it is broadened in the crossover regime at high temperatures. The T_{CEP} between first-order and crossover is indicated by arrows.

Figure 2 shows the field dependence of $d\rho_{xy}/dH$ at 2.25, 3.12, and 3.41 GPa. Clear maxima or minima were observed due to the PM–FM1 transition which is defined as H_C . As shown in Fig. 2(b), the transition width, indicated by the arrows, was estimated using the cross point of linear extrapolations. At 2.25 GPa, the transition width is unchanged below 12 K, although the anomaly changes from a maximum to a minimum. Above 12 K the anomalies become broader with the increase of transition width. Figure 2(d) shows the temperature variation of the transition width for various pressures. The transition width at low temperature is approximately 1 T and almost temperature independent, but it broadens when entering the crossover regime by increasing the temperature. The boundary between the first order transition and the crossover is indicated by arrows and allows us to determine T_{CEP} . The present results are in good agreement with the previous results obtained by $d\rho/dT$.⁴⁾ For example, the previous estimation by $d\rho/dT$ indicates T_{CEP} of 18 K at 1.82 GPa, while the present results obtained by $d\rho_{xy}/dH$ indicates T_{CEP} of 18 K at 1.80 GPa. As shown in Fig. 2, H_C increases and T_{CEP} de-

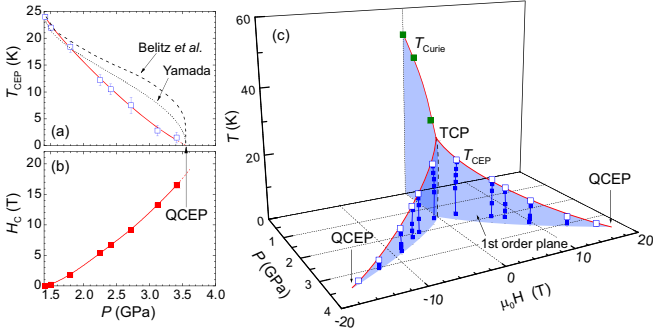


Fig. 3. (color online) Pressure dependence of (a) T_{CEP} and (b) H_C at low temperature. The QCEP is expected to be located at 3.5–3.6 GPa and 17–19 T from these plots. The dashed and dotted curves are the theoretical predictions by Belitz *et al.* and Yamada obtained using ($T_{\text{CEP}} = 24$ K, $P_{\text{QCEP}} = 3.55$ GPa, $H_{\text{QCEP}} = 18$ T).^{5,6)} (c): Three dimensional phase diagram of UGe₂.

creases with increasing pressure. At 3.12 GPa, T_{CEP} is approximately 3 K, and H_C is 13.2 T. At 3.41 GPa, H_C exceeds our maximum field of 16 T and only the beginning of the PM–FM1 transition is detected. We roughly estimated $T_{\text{CEP}} = 1.5 \pm 1$ K and $H_C = 16.5 \pm 0.5$ T at 3.41 GPa from comparison with data at other pressures.

Figures 3 show the pressure dependence of T_{CEP} and H_C at the lowest temperature. T_{CEP} decreases with pressure and the variation becomes weaker at higher pressure, while the slope of H_C becomes steeper. Figure 3(c) shows the three dimensional phase diagram drawn from the present results. T_{CEP} becomes zero at $P_{\text{QCEP}} \sim 3.5 - 3.6$ GPa and $H_{\text{QCEP}} \sim 17 - 19$ T from the extrapolations shown in Figs. 3(a) and (b).

Figure 4(a) shows temperature dependence of ρ_{xx} for several fields at 3.12 GPa close to H_{QCEP} where T_{CEP} and H_C are ~ 3 K and 13.2 T respectively. No clear anomaly was observed in ρ_{xx} at T_{CEP} within the experimental precision, although the anomaly is observed when T_{CEP} is high at lower pressures.⁴⁾ Figure 4(b) shows the T^2 plot of ρ_{xx} . At 0 T and 14.5 T, the Fermi liquid (FL) behavior of $\rho_{xx} = \rho_0 + AT^2$ is observed in a wide temperature range, while ρ_{xx} obeys the FL behavior below approximately $T_{\text{CEP}} \sim 3$ K at 12 T and 13.2 T. The A coefficient in the FL region is the largest at $H_C \sim 13.2$ T, indicative of an enhancement of quasiparticle mass on crossing H_C . The FL regime shrinks on approaching H_C , but careful analysis is required to discuss the origin of this behavior, since the residual resistivity ρ_0 , which is different between the PM phase and the FM1 phase, is expected to depend on temperature especially near H_C in a wide temperature range from the crossover region at high temperatures to the first order region at low temperatures.

Figure 4(c) shows the field dependences of ρ_{xy} at low temperatures and (d) of the A coefficient estimated from the field dependences of ρ_{xx} at two different temperatures by assuming the FL form of $\rho_{xx} = \rho_0 + AT^2$. ρ_{xy} shows a step-like anomaly at H_C up to 3.12 GPa, indicating that the drastic change in the FS is maintained up to the vicinity of the QCEP. On the other hand, the field dependence of A shows a large evolution toward the

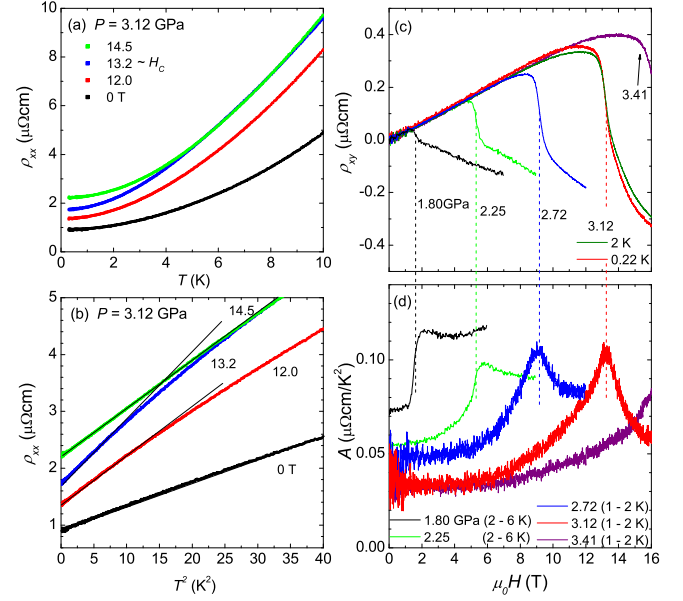


Fig. 4. (color online) (a) Temperature dependence of ρ_{xx} and (b) ρ_{xx} vs. T^2 at 3.12 GPa, where H_C is 13.2 T and T_{CEP} is estimated to be ~ 3 K from Fig. 2(d). Field dependence of (c) ρ_{xy} at low temperatures, and (d) A coefficient at several pressures. ρ_{xy} for 1.80 – 2.72 GPa was measured at 2 K, and 3.41 GPa is 0.22 K. The values of the A coefficient were obtained from the field dependence of ρ_{xx} at two denoted temperatures by assuming the FL form. The dotted lines indicate H_C at each pressure.

QCEP. At lower pressure with high T_{CEP} , A shows a step-like anomaly at H_C which is indicative of a first-order transition between two phases with different FS's and effective masses. This behavior is consistent with the previous report.¹⁵⁾ The step-like anomaly of A gradually changes into peak structures with increasing pressure. Both A at zero field and A at FM1 sufficiently above H_C decrease with increasing pressure, while A at H_C is almost pressure independent. It is apparent that the ratio $A(H_C)/A(H = 0)$ increases toward the QCEP. This strong modification in the mass enhancement may be driven by the continuous decrease of ΔM_0 on approaching QCEP at the benefit of a large increase in magnetic fluctuations. Similar enhancement of A under magnetic field has been observed in antiferromagnetic (AF) heavy fermion compounds such as CeRh₂Si₂ on crossing its first order metamagnetic transition²⁰⁾ or CeIn₃ and YbRh₂Si₂ on crossing its magnetic transition from AF to PM.^{21,22)} For the two cases, the magnetic transition is associated with FS changes. For example, in CeIn₃ it was proposed that it is associated to a Lifshitz instability,²³⁾ and in YbRh₂Si₂ the anomaly in Hall coefficient was interpreted as a collapse of the large FS.²⁴⁾

Studies on other FM materials such as ZrZn₂ suffer from the fact as the jump ΔM_0 is one order of magnitude smaller than that of UGe₂²⁾ the separation between P_{QCEP} and P_C seems very small and thus the FM wing has been only drawn schematically. The enhancement of the A coefficient at H_C is also observed in Sr₃Ru₂O₇²⁵⁾ which is considered to lie in the vicinity of FM QCEP. Unfortunately Sr₃Ru₂O₇ is already in PM ground state

and thus the validity of a dominant FM coupling is not obvious. It was stressed that an AF instability may play an important role²⁶⁾ with a possible duality between FM and AF tricriticality²⁷⁾ in a manner similar to the highly studied CeRu_2Si_2 family.^{28,29)} Finally, MnSi suffers from the complexity that due to its lack of an inversion symmetry, the ordered ground state at zero field is not a FM.¹⁾ UGe_2 is an excellent system with remarkable separation between P_{QCEP} and P_C .

Because of the large pressure and field extensions of the first-order plane, UGe_2 is a good test for theories of the FM phase diagram. Belitz *et al.* argued that long wavelength correlation effects can explain such a phase diagram with the wing structure.⁵⁾ In Belitz's model which gives T_{CEP} vs. H_C , we obtained the pressure dependence of T_{CEP} using the pressure dependence of H_C . However, as shown by the dashed line in Fig. 3(a), the quantitative agreement with our data is poor. Yamada explained the pressure dependence of T_{CEP} by taking into account the magnetoelastic coupling. In Yamada's model, we used $q(0) = 0.16$ and $\eta = 0.01$ as parameters in ref.,⁶⁾ where η connects to the magnetovolume coupling and the result has no large η dependence. However again the predicted T_{CEP} variation of $(P - P_{\text{QCEP}})^{1/2}$ does not seem to agree with our results. These discrepancies should inspire more refinements in theories. For example, it would be important to take into account the change of FS at the PM-FM1 transition which has been clearly detected by quantum oscillation experiments¹⁹⁾ and the present Hall resistivity measurements. The FS's of the phases FM2, FM1 and PM are quite different.^{13,19)} Thus an interesting problem is the role of FS change on the quantum metamagnetic transition. As pointed out in the hypothesis of a so-called Lifshitz transition, unconventional universality may occur at quantum metamagnetic transition.^{9,10)} The discrepancy between our results and the conventional description of the FM wing structure appears as the signature of a FS change at the PM-FM1 quantum singularity. It must be noticed that a strong dependence of T_{CEP} quite similar to that observed here was found in a band structure frame with a Van Hove singularity.⁸⁾ Thus FS instability appears a key ingredient to discuss its FM quantum instability and also in AF heavy fermion systems such as CeRh_2Si_2 , CeIn_3 , or YbRh_2Si_2 .

In summary, we have measured the (P, T, H) phase diagram of UGe_2 up to the vicinity of the QCEP using the Hall resistivity. The QCEP of PM - FM boundary is suggested to be located at 3.5 – 3.6 GPa and 17 – 19 T, where it is sufficiently apart from the FM critical point at zero field to be separately observed ($P_{\text{QCEP}}/P_C > 2$). A strong evolution of the field dependence of the A coefficient was observed from a strong first order regime up to the QCEP, while the Hall resistivity showed that the change in the FS at the PM-FM1 transition is maintained up to the vicinity of the QCEP.

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- 1) C. Pfleiderer, S. R. Julian, and G. G. Lonzarich: Nature (London) **414** (2001) 427.
- 2) M. Uhlarz, C. Pfleiderer, and S. M. Hayden: Phys. Rev. Lett. **93** (2004) 256404.
- 3) N. Kabeya, R. Iijima, E. Osaki, S. Ban, K. Imura, K. Deguchi, N. Aso, Y. Homma, Y. Shiokawa, and N. K. Sato: J. Phys. Conf. Ser. **200** (2010) 032028.
- 4) V. Taufour, D. Aoki, G. Knebel, and J. Flouquet: Phys. Rev. Lett. **105** (2010) 217201.
- 5) D. Belitz, T. R. Kirkpatrick, and J. Rollbühler: Phys. Rev. Lett. **94** (2005) 247205.
- 6) H. Yamada, Physica B **391** (2007) 42.
- 7) A. J. Millis, A. J. Schofield, G. G. Lonzarich, and S. A. Grigera: Phys. Rev. Lett. **88** (2002) 217204.
- 8) B. Binz and M. Sgrist: Europhys. Lett. **65** (2004) 816.
- 9) Y. Yamaji, T. Misawa, and M. Imada: J. Phys. Soc. Jpn. **76** (2007) 063702.
- 10) M. Imada, T. Misawa, and Y. Yamaji: J. Phys.: Condens. Matter **22** (2010) 164206.
- 11) S. S. Saxena, P. Agarwal, K. Ahilan, F. M. Grosche, R. K. W. Haselwimmer, M. J. Steiner, E. Pugh, I. R. Walker, S. R. Julian, P. Monthoux, G. G. Lonzarich, A. Huxley, I. Sheikin, D. Braithwaite, and J. Flouquet: Nature (London) **406** (2000) 587.
- 12) C. Pfleiderer and A. D. Huxley, Phys. Rev. Lett. **89** (2002) 147005.
- 13) Y. Haga, M. Nakashima, R. Settai, S. Ikeda, T. Okubo, S. Araki, T. C. Kobayashi, N. Tateiwa, and Y. Ōnuki: J. Phys.: Condens. Matter **14** (2002) L125.
- 14) T. C. Kobayashi, K. Hanazono, N. Tateiwa, K. Amaya, Y. Haga, R. Settai, and Y. Ōnuki: J. Phys.: Condens. Matter **14** (2002) 10779.
- 15) T. Terashima, K. Enomoto, T. Konoike, T. Matsumoto, S. Uji, N. Kimura, M. Endo, T. Komatsubara, H. Aoki, and K. Maezawa: Phys. Rev. B **73** (2006) 140406(R).
- 16) T. C. Kobayashi, H. Hidaka, H. Kotegawa, K. Fujiwara, and M. I. Eremets: Rev. Sci. Instrum. **78** (2007) 023909.
- 17) K. Murata, K. Yokogawa, H. Yoshino, S. Klotz, P. Munsch, A. Irizawa, M. Nishiyama, K. Iizuka, T. Nanba, T. Okada, Y. Shiraga, and S. Aoyama: Rev. Sci. Instrum. **79** (2008) 085101.
- 18) V. H. Tran, S. Paschen, R. Troć, M. Baenitz, and F. Steglich: Phys. Rev. B **69** (2004) 195314.
- 19) R. Settai, M. Nakashima, S. Araki, Y. Haga, T. C. Kobayashi, N. Tateiwa, H. Yamagami, and Y. Ōnuki: J. Phys.: Condens. Matter **14** (2002) L29.
- 20) W. Knafo, D. Aoki, D. Vignolles, B. Vignolle, Y. Klein, C. Jaudet, A. Villaume, C. Proust, and J. Flouquet: Phys. Rev. B **81** (2010) 094403.
- 21) A. V. Silhanek, Takao Ebihara, N. Harrison, M. Jaime, Koji Tezuka, V. Fanelli, and C. D. Batista: Phys. Rev. Lett. **96** (2006) 206401.
- 22) P. Gegenwart, J. Custers, C. Geibel, K. Neumaier, T. Tayama, K. Tenya, O. Trovarelli, and F. Steglich: Phys. Rev. Lett. **89** (2002) 056402.
- 23) L. P. Gor'kov and P. D. Grigoriev: Phys. Rev. B **73** (2006) 060401(R).
- 24) S. Paschen, T. Lühmann, S. Wirth, P. Gegenwart, O. Trovarelli, C. Geibel, F. Steglich, P. Coleman and Q. Si: Nature **432** (2004) 881.
- 25) S. A. Grigera, R. S. Perry, A. J. Schofield, M. Chiao, S. R. Julian, G. G. Lonzarich, S. I. Ikeda, Y. Maeno, A. J. Millis, A. P. Mackenzie: Science **294** (2001) 329.
- 26) K. Kitagawa, K. Ishida, R. S. Perry, T. Tayama, T. Sakakibara, and Y. Maeno: Phys. Rev. Lett. **95** (2005) 127001.

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- 27) T. Misawa, Y. Yamaji, and M. Imada: J. Phys. Soc. Jpn. **78** (2009) 084707.
- 28) J. Flouquet, in: “*Progress in Low Temperature Physics*”, Ed. by W.P. Halperin, (Elsevier, Amsterdam, 2006) p. 139.
- 29) D. Aoki, C. Paulsen, T. D. Matsuda, L. Malone, G. Knebel, P. Haen, P. Lejay, R. Settai, Y. Ōnuki, and J. Flouquet: J. Phys. Soc. Jpn. **80** (2011) 053702.